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Nitric Oxide Destruction in the Fuel-Bed Burning Regime of Spreader Stokers

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16. ABSTRACT The article gives results of an experimental study of nitric oxide (NO) destruction in the fuel-bed of a coal-fired spreader stoker. NO was injected into the coal bed and freeboard flame zone under varying local stoichiometries to determine the fate of NO in the primary combustion zone. In general, a high capacity for NO reduction was observed, with bed-phase reduction contributing most significantly to the overall mechanism. Fractional survival of injected NO, as well as fuel nitrogen conversion to exhaust NO, decreased with decreasing bed-region stoichiometry. Staged firing also decreased NO survival by increasing residence time in the fuel-rich primary combustion zone. The modest nitrogen oxides (NOx) reductions achievable with Flue Gas Recirculation (FGR), a practical NOx control technique, are due in part to the more than 50% destruction of NO in the recycled flue gases.

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Nitric Oxide Destruction in the Fuel-Bed Burning Regime of Spreader Stokers

An experimental study of nitric oxide (NO) destruction in the fuel bed of a coal-fired spreader stoker was conducted. NO was injected into the coal bed and freeboard flame zone under varying local stoichiometries to determine the fate of NO in the primary combustion zone. In general, a high capacity for NO reduction was observed, with bed-phase reduction contributing most significantly to the overall mechanism. Fractional survival of injected NO, as well as fuel nitrogen conversion to exhaust NO, decreased with decreasing bed-region stoichiometry. Staged firing also decreased NO survival by increasing residence time in the fuel-rich primary combustion zone. The modest nitrogen oxides (NO_x) reductions achievable with flue gas recirculation (FGR), a practical NO_x control technique, are due in part to the more than 50% destruction of NO in the recycled flue gases.

The reduction of nitrogen oxides (NO_x) emissions from coal-fired spreader stokers has been partially demonstrated in pilot- and large-scale testing (Giammar et al., 1979; Langsjeon et al., 1981). NO_x control schemes have been primarily limited to modifications in the operating procedures of existing equipment. Quartucy et al. (1986) tested flue gas recirculation (FGR) in five full-scale stoker boilers and obtained only small decreases in NO_x emissions. At all sites the fuel beds were fired with high excess air levels, and the flue gas was recycled almost entirely above the beds. Maloney (1985) demonstrated at full scale that FGR can significantly enhance fuel burnout (improve carbon utilization) and lower NO_x emissions. Recent studies (Starley et al., 1983, 1985a,b) have helped to characterize the relative effects of combustion parameters on NO formation in fixed-bed coal firing. These studies were principally concerned with determining the influence of local combustion environment on the fate of nitrogenous species. The work demonstrated that bed-region (first-stage) stoichiometry controlled fuel nitrogen speciation; locally fuel-rich conditions favored the formation of non-oxidized intermediates (NH_3 and HCN). Second-stage oxidation of the reduced nitrogen species was inherently low due to reduction reactions favoring N_2 formation. Greatly reduced exhaust NO emissions resulted from staged burning.

This brief paper describes the results of an experimental investigation of the fate of NO formed in the fuel-bed burning regime of spreader stokers. The work was further intended to assess the relative importance of gas-phase homogeneous reactions and heterogeneous gas–solid reactions in NO reduction. (These reactions are important because they control the fate of both NO formed low in the bed and NO present in recycled flue gases.) Bed-region (first-stage) and overall furnace stoichiometries were used in data correlation. Stoichiometry (SR) was defined as the ratio of available combustion air to the air required for complete combustion to theoretical products. SR is used here as an air-to-fuel ratio; a fuel-rich condition corresponds to an SR of less than 1.0.

An experimental fixed-bed furnace, shown in Figure 1, was utilized for simulating the fuel-bed burning regime in coal-fired spreader stokers (Starley et al., 1985b). The combustor was designed for firing rates comparable to

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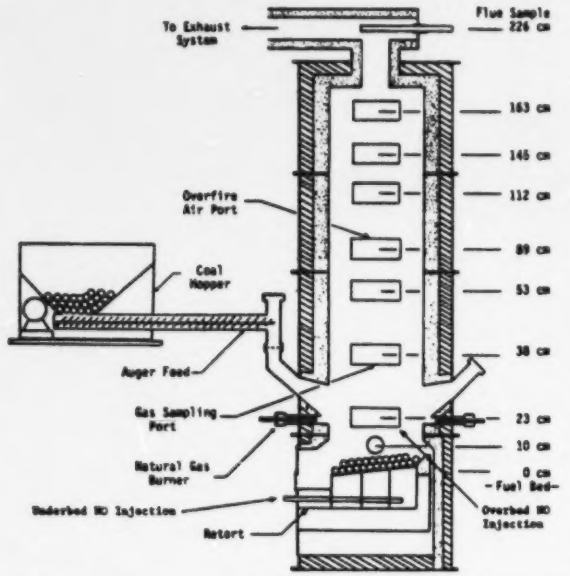

Figure 1. Laboratory spreader stoker furnace configured for NO injection experiments.

commercial traveling grate, spreader stokers (1.6–1.9 MW/m^2), with a total bed surface area of 700 cm^2. Several sampling ports and windows were available for monitoring local combustion conditions.

Coal was fed to the furnace continuously from a storage hopper by a variable-speed auger. Coal fines were removed from the feed to provide a fuel particle size distribution ranging from 6 to 32 mm. This configuration allowed the formation and sustained burning of a 10–15-cm deep fuel bed.

Primary combustion air (underfire air, UFA) was supplied through three independent underbed plenums. Secondary combustion air (overfire air, OFA) was introduced to the overbed region as high velocity jets (approximately 30.5 m/s), injected normal to the axial (vertical) flow of combustion gases, and provided turbulent mixing in the overfire region. All air streams were metered with rotameters and supplied at ambient temperature.

Exhaust gas samples were withdrawn from the flue through a water-cooled, stainless steel probe. Measurements were made continuously and included chemiluminescence NO/NO_2, paramagnetic O_2, and NDIR CO and CO_2. Combustion zone gas temperatures were monitored with a suction pyrometer at approximately 10 cm above the fuel bed. A complete description of the experimental facility and operation has been reported elsewhere (Starley et al., 1985b).

Experimental Results and Discussion

An experimental investigation of the influence of bed-region stoichiometry on the fate of NO was conducted. This work was an extension of previous studies on the fate of fuel nitrogen in stoker-coal-fired furnaces (Starley et al., 1985b). Two sets of experiments were performed in which pure NO was injected into the furnace along with the combustion air. NO was selected as the trace species (dopant) in order to follow the fate of NO formed in the fuel bed and conveyed through the active combustion region. In the initial experiments, pure NO was injected into the underfire (primary) air, and the change in NO emissions was monitored in the exhaust gases. The NO was precisely metered into the combustion air through a critical orifice.

Table I. Utah Coal Analysis

	as received, %	dry basis, %
proximate anal.		
moisture	6.20	
ash	9.20	9.81
volatile matter	37.62	40.10
fixed carbon	46.98	50.09
ultimate anal.		
moisture	6.20	
carbon	66.93	71.35
hydrogen	4.65	4.96
nitrogen	1.11	1.18
sulfur	0.33	0.35
oxygen	11.58	12.35
ash	9.20	9.81
fuel constituent (%, daf)[a]		
nitrogen	1.31	
calorific value (as received)	20 828 kg cal/kg 11 571 Btu/(lb m)	
calorific value (wet, mmf)[b]	22 937 kg cal/kg 12 743 Btu/(lb m)	
ASTM rank[c]	HVC bituminous	

[a] daf: dry, ash free. [b] mmf: mineral matter free. [c] HVC: high volatile carbon.

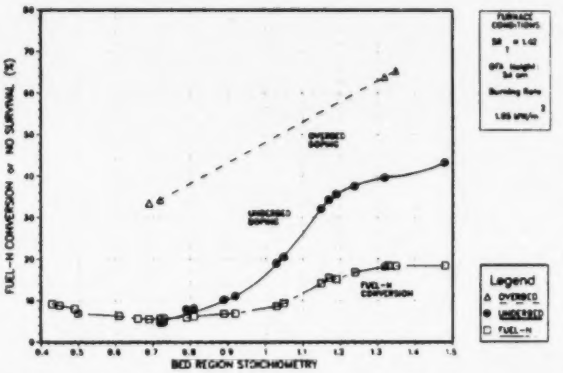

Figure 2. Fuel nitrogen conversion to exhaust NO and fractional survival of injected NO.

Additional experiments were performed to assess the extent of gas-phase reactions in the flame zone above the fuel bed. In these experiments, NO was injected at approximately 3 cm above the bed into the first-stage combustion gases, and the change in exhaust NO emissions was monitored. In all experiments the overall furnace stoichiometry remained constant at approximately 1.42 (42% excess air, 6% excess exhaust oxygen), while the bed-region stoichiometry was varied by changing the overfire-to-underfire air ratio. A high-volatile Utah bituminous coal was fired in these studies (Table I).

Figure 2 illustrates fuel nitrogen conversion to exhaust NO (squares) as a function of bed-region stoichiometry for the coal alone. In general the data show that fuel nitrogen conversion is inherently low for the fuel-bed burning regime of spreader stokers. Under excess air burning, a maximum of approximately 16.5% (580 ppm at 0% excess O_2) was obtained. A minimum conversion of approximately 6.5% (210 ppm at 0% excess O_2) was achieved at a fuel-rich bed stoichiometry of 0.65. Under heavily staged combustion, conversion increased to nearly 9% (275 ppm at 0% excess O_2). This general trend in the influence of primary combustion zone stoichiometry on fuel nitrogen conversion has been observed by several investigators for a variety of coal-fired systems (Chen et al., 1982; Munro, 1982; Rees et al., 1981; Starley et al., 1985a,b).

Table II. Injected NO Survival

	bed stoichiometry	
	0.72	1.32
NO survival to exhaust:		
overbed doping, %	34.2	63.8
underbed doping, %	4.5	39.6
local NO survival		
flame-zone phase, %	34.2	63.8
fuel-bed phase, %	13.3	62.4
flame-zone conditions		
gas temp, °C[b]	1450	1510
nitrogenous species[a-c]		
NO, ppm	31	497
NH_3, ppm	348	22
HCN, ppm	227	4
furnace residence times		
bed region, s	0.10	0.05
flame zone, s	0.58	0.31

[a] Nitrogenous species measured without NO doping occurring. [b] 10 cm above bed. [c] Dry, at $SR_B = 1.0$.

Figure 2 further illustrates fractional survival of injected NO. Solid circles refer to periods of NO injection directly into the fuel bed, and open triangles correspond to doping in the freeboard flame zone directly above the bed.

The data show that NO was readily reduced under all stoichiometric conditions. Even under high excess air firing ($SR_B = 1.48$), only 43% of the NO injected through the fuel bed survived and reached the exhaust. Dopant survival decreased with decreasing bed-region stoichiometry, and a minimum of approximately 4% NO retention was observed at a stoichiometry of 0.72.

Doping over the fuel bed (but beneath the overfire air injectors) was conducted to assess the effects of gas-phase flame zone stoichiometry on NO reduction. Overbed doping resulted in a 64% NO survival under excess air burning and a 34% retention under fuel-rich conditions.

Comparison of the coal nitrogen conversion results and the underbed doping data shown in Figure 2 indicates that, under fuel-rich conditions, the source and form of the nitrogen have relatively little influence on the fraction which is ultimately emitted as NO. Nitrogen which enters the bed (from either the fuel or recycled flue gas) is apparently rapidly assimilated into the overall XN pool (NHi, CN, and NO species).

Table II gives a comparison of dopant survival through the fuel bed and the flame zone under excess air ($SR_B = 1.32$) and fuel-rich ($SR_B = 0.72$) conditions. In-flame, gas-phase concentrations of nitrogenous species (at 10 cm above the fuel bed) measured under coal firing (not during doping periods) are also reported, as well as local gas temperatures.

Fractional NO retention was calculated separately for the bed and gas-phase (freeboard) flame zone. Gas-phase survival was first calculated directly from the overbed NO injection experiments; local bed-phase NO survival was then determined by material balance using overall NO survival data from the underbed injection cases.

Under staged firing, approximately 13% of the injected NO survived passage through the fuel bed, and of the NO remaining, only 34% NO survived the fuel-rich flame zone above the bed, producing an overall survival ratio of only 4.5%. When the coal bed was fired with excess air, approximately 62% of the dopant survived the bed-phase combustion region, which was roughly equivalent to the NO retention observed in the flame zone, producing an overall survival ratio of approximately 39%.

These data demonstrate that the fuel bed is extremely effective at reducing NO, particularly under fuel-rich conditions. This reduction is almost certainly due to the existence of local fuel-rich regimes in the near field of the particles and to heterogeneous reactions with solid-phase carbon. The relative contributions of homogeneous and gas–solid reduction reactions in the fuel bed cannot be separated; however, the influence of local stoichiometry is well established.

In the freeboard flame zone above the fuel bed, the dominant NO reduction mechanism is probably homogeneous. Levy et al. (1981) and DeSoete (1980) demonstrated that NO reduction on char surfaces could influence overall NO formation in pulverized coal combustion. Glass and Wendt (1983) studied nitrogenous species reduction for a Utah coal and its char during staged pulverized coal firing. Their data suggest bulk NO reduction rates of approximately 1×10^{-3} mol of NO/s through the primary combustion zone at gas temperatures ranging from 1880 to 2080 °C, which compare favorably with bulk rates estimated from the global measurements of this study (1.4×10^{-4} mol of NO/s) at 1450–1500 °C. Activation energies and reaction rates were not obtained in this work. However, bulk NO reduction was within general agreement and at least directionally correct (when considering the temperature difference) in relation to Glass and Wendt's data. They concluded that heterogeneous reduction reactions provided only a minor contribution to overall NO destruction and that gas-phase reactions determined the fate of nitrogen intermediates. Particulate carryover from the fuel bed in stokers leads to much lower entrained solids densities than observed in pulverized coal flames. Consequently, heterogeneous NO reduction in the overbed flame zone should be even less significant in stoker-coal-fired systems; homogeneous reactions should control the fate of NO in the freeboard flame zone.

Conclusions

The fuel-bed burning regime of spreader stokers was demonstrated to have a high capacity for reducing NO under all stoichiometric conditions. The fractional survival of injected NO decreased with decreasing bed-region stoichiometry. Staged firing decreased the overall NO survival by increasing the residence time in the fuel-rich primary combustion region (bed-phase and freeboard flame zone). Fuel nitrogen conversion to exhaust NO was also observed to decrease with decreasing bed-region stoichiometry, resulting in substantially decreased NO_x emissions.

Flue gas recirculation should be a viable method of achieving modest NO_x reductions in stoker furnaces. The current commercial practice is to recycle the flue gases above the stoker bed; under normal operating conditions, this should produce a direct reduction in exhaust emission because approximately one-third of the NO contained in the recycled gases will be destroyed. If the flue gases were recycled through the coal bed itself, then an additional reduction would be achieved; approximately 60% of the recycled NO would be destroyed. (Flue gas recirculation may also produce additional benefits and/or problems not defined here because of the altered mixing and overall thermal profiles.) Where possible, stoker firing should be staged to reduce the primary NO emission levels and to maximize the capacity for first-stage NO reduction.

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